

U.S. Patent Application Serial No. 10/019,287
Amendment filed November 14, 2005
Reply to OA dated June 15, 2005

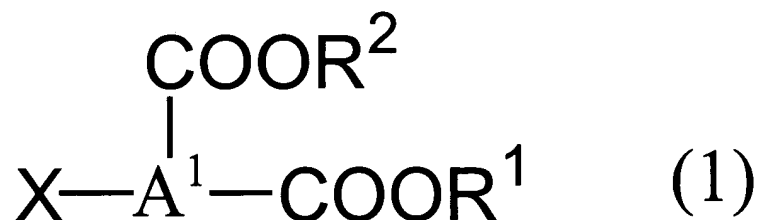
AMENDMENTS TO THE CLAIMS:

Please cancel claims 1-5, 12-18, and 31-33 without prejudice or disclaimer, and amend claims 11, 19, 20, 30, 34, 44, 45, 64, 65, 76, 86, and 87, as follows. This listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

Claims 1-5 (canceled).

Claim 6 (Previously presented): A process for preparing an alicyclic dicarboxylic acid diester represented by the formula (1)



wherein A¹ represents a cyclohexane ring or cyclohexene ring, X is a hydrogen atom or methyl, R¹ and R² are the same or different and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms; and having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,

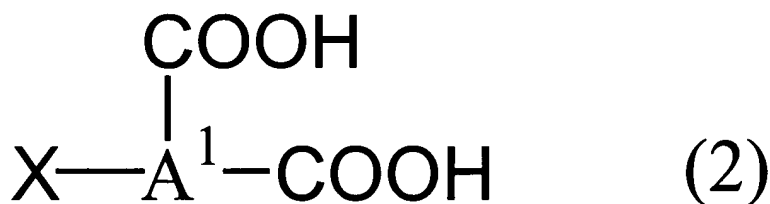
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- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of

(i) subjecting

- a) an alicyclic dicarboxylic acid represented by the formula (2)



wherein A^1 and X are as defined above, or an anhydride thereof, and

- b) an aliphatic monohydric alcohol having 1 to 18 carbon atoms or an alicyclic monohydric alcohol having 3 to 10 carbon atoms each having a peroxide value of 1.0 meq/kg or less to esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst, or subjecting

a') an alicyclic dicarboxylic acid diester represented by the formula (3)



wherein A¹ and X are as defined above, R³ and R⁴ are the same or different and each is a branched-chain alkyl group having 3 or 4 carbon atoms or a straight-chain alkyl group having 1 to 4 carbon atoms, and

b') an aliphatic monohydric alcohol of 5 to 18 carbon atoms or an alicyclic monohydric alcohol of 3 to 10 carbon atoms each having a peroxide value of 1.0 meq/kg or less to ester interchange reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst, to thereby obtain a reaction mixture containing the diester represented by the formula (1),

(ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the diester in a crude form,

(iii) neutralizing the crude diester obtained in step (ii) and washing the neutralized crude diester with water,

(iv) purifying the crude diester neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents, and

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(v) dehydrating the diester purified in step (iv).

Claim 7 (original): The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein said alcohols under b) or b') used in step (i) has a carbonyl value of 15 or less.

Claim 8 (original): The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein the esterification reaction or the ester interchange reaction in step (i) is carried out in an inert gas atmosphere or in an inert gas stream.

Claim 9 (original): The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein the esterification reaction or the ester interchange reaction in step (i) is carried out in the presence of a sulfur-free and phosphorus-free catalyst, the catalyst being selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃ -C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 10 (original): The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein in step (iii), the neutralization is carried out until the crude diester has

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a total acid number of 0.05 mgKOH/g or less after being washed with water, and the crude diester is washed with water until the pH of the washings used for the washing becomes neutral.

Claim 11 (Currently amended): The process for preparing the alicyclic dicarboxylic acid diester according to claim 6, wherein the treatment with adsorbents in step (iv) is carried out using 2 to 4 the adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

Claims 12-18 (canceled).

Claim 19 (Currently amended): A process for preparing an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester or an ester mixture, the alicyclic ~~or aromatic~~ adjacent adjacent dicarboxylic acid mixed diester being represented by the formula (4)

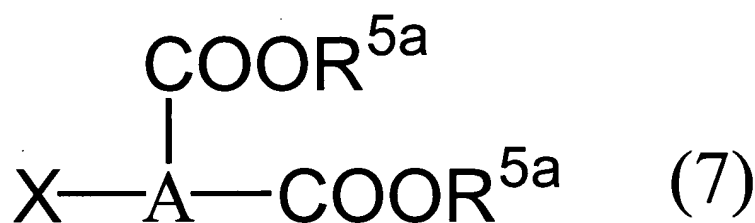


wherein A represents a cyclohexane ring[[,]] or a cyclohexene ring ~~or a benzene ring~~, X is a

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hydrogen atom or methyl, R^5 and R^6 are different from each other and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms, and the group $-\text{COOR}^5$ and the group $-\text{COOR}^6$ are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring or benzene ring represented by A; and said ester mixture being a mixture of

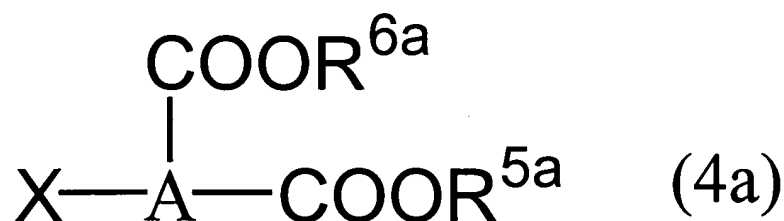
(1) an alicyclic or aromatic adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7)



wherein A and X are as defined in the formula (4), and R^{5a} represents a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and the two $-\text{COOR}^{5a}$ groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring or benzene ring represented by A,

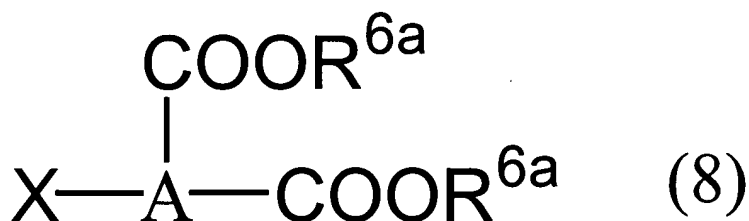
(2) an alicyclic or aromatic adjacent dicarboxylic acid mixed diester represented by the

formula (4a)



wherein A and X are as defined in the formula (7), and R^{5a} and R^{6a} are different from each other and R^{5a} is as defined in the formula (7), and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the group -COOR^{5a} and the group -COOR^{6a} are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring or benzene ring represented by A, and

(3) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl)ester represented by the formula (8)



wherein A, X and R^{6a} are as defined in the formula (4a), and the two -COOR^{6a} groups are the same

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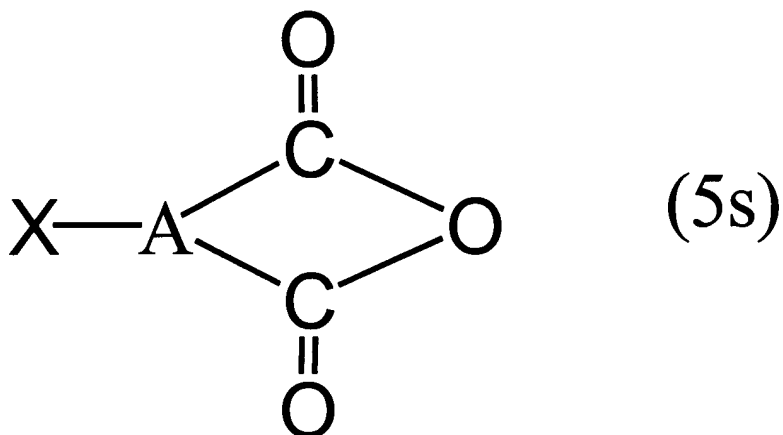
and attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or~~
~~benzene ring~~ represented by A, and

the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester or the ester mixture having
the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of

- (i) (a) subjecting an alicyclic ~~or aromatic~~ adjacent dicarboxylic anhydride
represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A, X and R⁵ are as defined above, and the group –COOR⁵ and the group –COOH are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A,

- (b) subjecting the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst to thereby give a reaction mixture containing said ester mixture of (1) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), (2) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a), and (3) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
- (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents, and

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- (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having the properties 1) to 9).

Claim 20 (Currently amended): The process according to claim 19, wherein the ester mixture contains the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a) under (2) in a proportion of 100, the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7) under (1) in a proportion of 5-300, and the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8) under (3) in a proportion of 7-500, wherein the proportions are expressed in terms of area ratio as determined from a gas chromatogram of the ester mixture.

Claim 21 (original): The process according to claim 19 wherein the ester mixture is a mixture of an alicyclic adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), an alicyclic adjacent dicarboxylic acid mixed diester represented by the formula (4a) and an alicyclic adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8), the ester mixture having a trans isomer/cis isomer ratio of 0/100 to 80/20 (by area % as determined by gas chromatography).

Claim 22 (previously presented): The process according to claim 19, wherein the monohydric alcohol of 1 to 5 carbon atoms (P) constituting said alcohol component 1, is an alcohol

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composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (Q) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group, and

wherein the monohydric alcohol of 1 to 5 carbon atoms (S) constituting said alcohol component 2 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (T) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group.

Claim 23 (previously presented): The process according to claim 19, wherein said alcohol component 1 is a monohydric alcohol of 1 to 5 carbon atoms and said alcohol component 2 is a monohydric alcohol of 6 to 18 carbon atoms.

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Claim 24 (previously presented): The process according to claim 19, wherein said alcohol component 1 and said alcohol component 2 have a peroxide value of 1.0 meq/kg or less.

Claim 25 (previously presented): The process according to claim 24, wherein said alcohol component 1 and said alcohol component 2 further have a carbonyl value of 15 or less.

Claim 26 (original): The process according to claim 19 wherein said esterification reactions in steps (a) and (b) of step (i) are carried out in an inert gas atmosphere or in an inert gas stream.

Claim 27 (original): The process according to claim 19 wherein said reaction in step (a) of step (i) is carried out in the absence of a catalyst and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 28 (original): The process according to claim 19 wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is 10 to 90 mole%, relative to the total amount [(P)+(Q)+(S)+(T)] of alcohol component 1 [(P)+(Q)] used in the first stage esterification reaction

and alcohol component 2 [(S)+(T)] used in the second-stage esterification reaction, and

- 1) the whole amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in an amount of 50 mole% relative to the total amount [(P)+(Q)+(S)+(T)] in the first-stage esterification reaction and the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S) in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is more than 50 mole% and not more than 90 mole% relative to the total amount [(P)+(Q)+(S)+(T)].

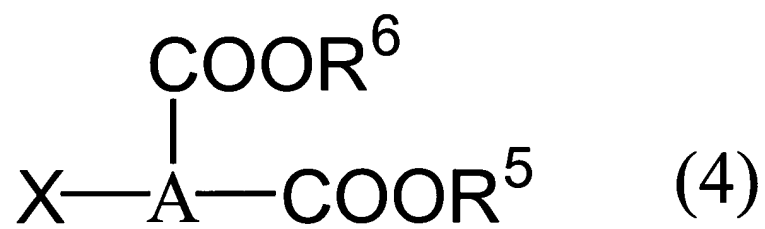
Claim 29 (original): The process according to claim 19 wherein in step (iii), the neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water until the pH of the washings used for the washing becomes neutral.

Claim 30 (Currently amended): The process according to claim 19, wherein the treatment with adsorbents in step (iv) is carried out using 2 to 4 the adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

Claims 31-33 (canceled).

Claim 34 (Currently amended): A process for preparing an ester mixture, the ester mixture being a mixture of:

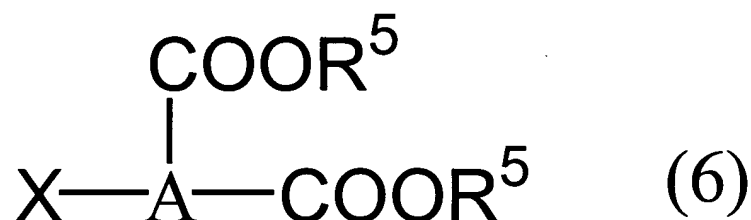
(1) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4)



wherein A represents a cyclohexane ring[[,]] or a cyclohexene ring ~~or a benzene ring~~, X is a hydrogen atom or methyl, R⁵ and R⁶ are different from each other and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a

straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms, and the group -COOR^5 and the group -COOR^6 are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

(2) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid diester represented by the formula (6)



wherein A, X and R^5 are as defined above, and two R^5 are the same, and the two -COOR^5 groups are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

the ester mixture having the following properties:

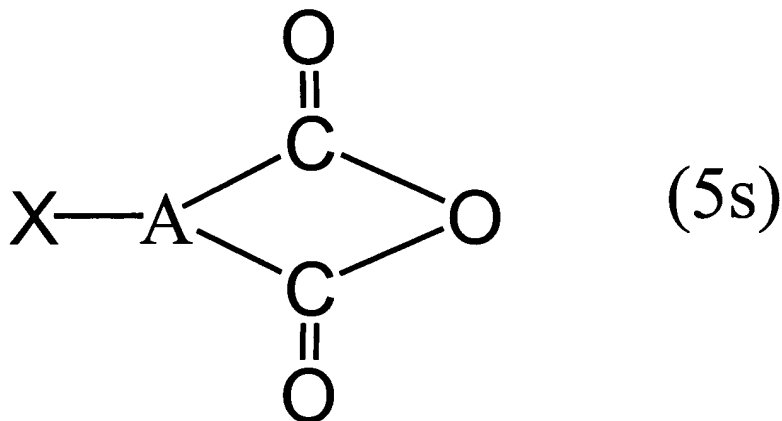
- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \bullet \text{cm}$ or more,

8) a hydroxyl value of 3 mgKOH/g or less, and

9) a water content of 100 ppm or less,

the process comprising the steps of:

- (i) (a) subjecting an alicyclic ~~or aromatic~~ adjacent dicarboxylic anhydride represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A, X and R⁵ are as defined above, and the group –COOR⁵ and the group –COOH are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] ~~or cyclohexene ring or benzene ring~~ represented by A,

- (b) subjecting the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst to thereby give a reaction mixture containing said ester mixture of the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4) and the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid diester represented by the formula (6),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
- (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents, and
- (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having

the properties 1) to 9).

Claim 35 (Previously presented): The process according to claim 34, wherein the monohydric alcohol of 1 to 5 carbon atoms (P) constituting said alcohol component 1 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (Q) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group, and

wherein the monohydric alcohol of 1 to 5 carbon atoms (S) constituting said alcohol component 2 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (T) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group.

Claim 36 (Previously presented): The process according to claim 34, wherein said alcohol component 1 is a monohydric alcohol of 1 to 5 carbon atoms and said alcohol component 2 is a monohydric alcohol of 6 to 18 carbon atoms.

Claim 37 (Previously presented): The process according to claim 34, wherein said alcohol component 1 and said alcohol component 2 have a peroxide value of 1.0 meq/kg or less.

Claim 38 (Previously presented): The process according to claim 37, wherein said alcohol component 1 and said alcohol component 2 further have a carbonyl value of 15 or less.

Claim 39 (Previously presented): The process according to claim 34, wherein said esterification reactions in steps (a) and (b) of step (i) are carried out in an inert gas atmosphere or in an inert gas stream.

Claim 40 (Previously presented): The process according to claim 34, wherein said reaction in step (a) of step (i) is carried out in the absence of a catalyst and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide,

lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 41 (Previously presented): The process according to claim 34, wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is 10 to 90 mole%, relative to the total amount [(P)+(Q)+(S)+(T)] of alcohol component 1 [(P)+(Q)] used in the first stage esterification reaction and alcohol component 2 [(S)+(T)] used in the second-stage esterification reaction, and

- 1) the entire amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in an amount of 50 mole% relative to the total amount [(P)+(Q)+(S)+(T)] in the first-stage esterification reaction and the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S) in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is more than 50 mole% and not more than 90 mole% relative to the total amount [(P)+(Q)+(S)+(T)].

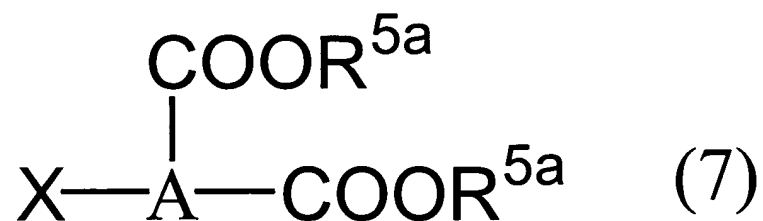
Claim 42 (Previously presented): The process according to claim 34, wherein in step (iii),

said neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water until the pH of the waste water becomes neutral.

Claim 43 (Previously presented): The process according to claim 34, wherein said treatment with adsorbents in step (iv) is carried out using 2 to 4 adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

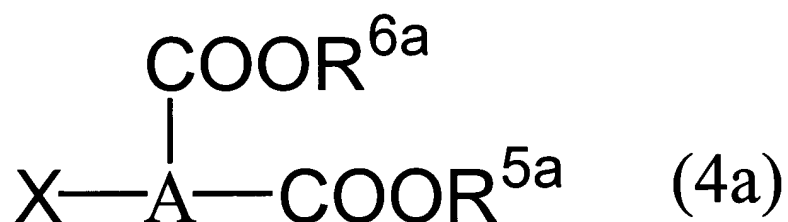
Claim 44 (Currently amended): A process for preparing an ester mixture, the ester mixture being a mixture of

(1) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7)



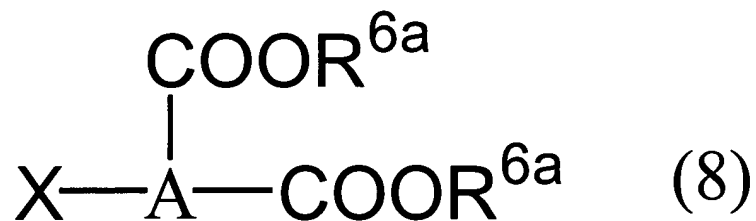
wherein A represents a cyclohexane ring~~[[,]]~~ or a cyclohexene ring ~~or a benzene ring~~, X is a hydrogen atom or methyl, and R^{5a} represents a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and the two -COOR^{5a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring~~[[,]]~~ or cyclohexene ring ~~or benzene ring~~ represented by A,

(2) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a)



wherein A and X are as defined in the formula (7), and R^{5a} and R^{6a} are different from each other and R^{5a} is as defined in the formula (7), and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the group -COOR^{5a} and the group -COOR^{6a} are attached to two adjacent carbon atoms of the cyclohexane ring~~[[,]]~~ or cyclohexene ring ~~or benzene ring~~ represented by A, and

(3) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl)ester represented by the formula (8)



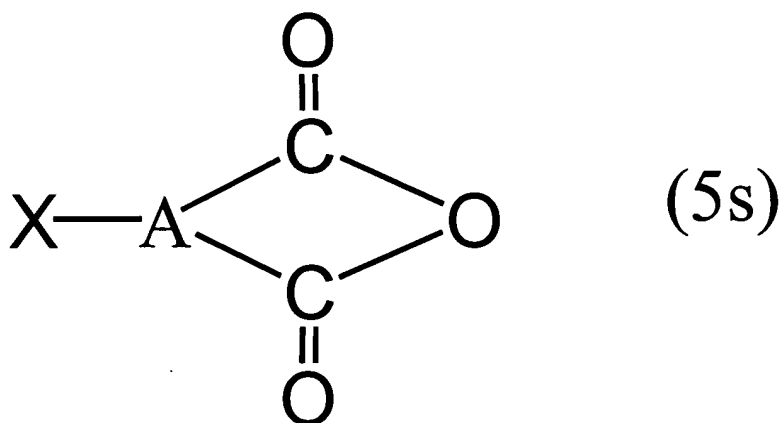
wherein A, X and R^{6a} are as defined in the formula (4a), and the two -COOR^{6a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

the ester mixture having the following properties:

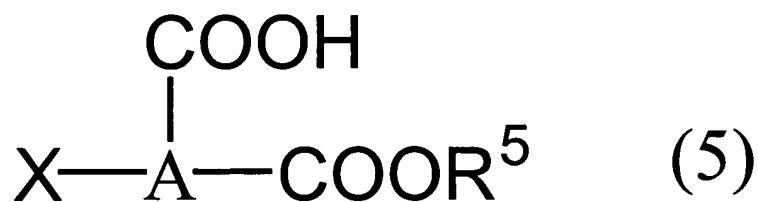
- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of:

- (i) (a) subjecting an alicyclic ~~or aromatic~~ adjacent dicarboxylic anhydride represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A and X are as defined above, and R⁵ is a branched-chain alkyl group having 3 to 18 carbon

atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms, and the group $-\text{COOR}^5$ and the group $-\text{COOH}$ are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring or ~~benzene ring~~ represented by A,

- (b) subjecting the alicyclic or ~~aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst to thereby give a reaction mixture containing said ester mixture of (1) the alicyclic or ~~aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), (2) the alicyclic or ~~aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a), and (3) the alicyclic or ~~aromatic~~ adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,

- (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents, and
- (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having the properties 1) to 9).

Claim 45 (Currently amended): The process according to claim 44, wherein the ester mixture contains the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by formula (4a) under (2) in a proportion of 100, the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by formula (7) under (1) in a proportion of 5-300, and the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl) ester represented by formula (8) under (3) in a proportion of 7-500, wherein the proportions are expressed in terms of area ratio as determined from a gas chromatogram of the ester mixture.

Claim 46 (Previously presented): The process according to claim 44, wherein the ester mixture is a mixture of an alicyclic adjacent dicarboxylic acid di(lower alkyl) ester represented by formula (7), an alicyclic adjacent dicarboxylic acid mixed diester represented by formula (4a) and an alicyclic adjacent dicarboxylic acid di(higher alkyl) ester represented by formula (8), the ester mixture having a trans isomer/cis isomer ratio of 0/100 to 80/20, wherein the ratios are expressed in terms of area %, as determined by gas chromatography.

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Claim 47 (Previously presented): The process according to claim 44, wherein the monohydric alcohol of 1 to 5 carbon atoms (P) constituting said alcohol component 1 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (Q) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group, and

wherein the monohydric alcohol of 1 to 5 carbon atoms (S) constituting said alcohol component 2 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (T) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group.

Claim 48 (Previously presented): The process according to claim 44, wherein said alcohol component 1 is a monohydric alcohol of 1 to 5 carbon atoms and said alcohol component 2 is a

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monohydric alcohol of 6 to 18 carbon atoms.

Claim 49 (Previously presented): The process according to claim 44, wherein said alcohol component 1 and said alcohol component 2 have a peroxide value of 1.0 meq/kg or less.

Claim 50 (Previously presented): The process according to claim 49, wherein said alcohol component 1 and said alcohol component 2 further have a carbonyl value of 15 or less.

Claim 51 (Previously presented): The process according to claim 44, wherein said esterification reactions in steps (a) and (b) of step (i) are carried out in an inert gas atmosphere or in an inert gas stream.

Claim 52 (Previously presented): The process according to claim 44, wherein said reaction in step (a) of step (i) is carried out in the absence of a catalyst, and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 53 (Previously presented): The process according to claim 44, wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms $[(P)+(S)]$ is 10 to 90 mole%, relative to the total amount $[(P)+(Q)+(S)+(T)]$ of alcohol component 1 $[(P)+(Q)]$ used in the first stage esterification reaction and alcohol component 2 $[(S)+(T)]$ used in the second-stage esterification reaction, and

- 1) the entire amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms $[(P)+(S)]$ is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in an amount of 50 mole% relative to the total amount $[(P)+(Q)+(S)+(T)]$ in the first-stage esterification reaction and the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S) in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms $[(P)+(S)]$ is more than 50 mole% and not more than 90 mole% relative to the total amount $[(P)+(Q)+(S)+(T)]$.

Claim 54 (Previously presented): The process according to claim 44, wherein in step (iii), said neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water until the pH of the waste water becomes neutral.

Claim 55 (Previously presented): The process according to claim 44, wherein said treatment with adsorbents in step (iv) is carried out using 2 to 4 adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

Claim 56 (Previously presented): The process according to claim 34, wherein R^5 is a straight-chain alkyl group having 1 to 5 carbon atoms or a branched-chain alkyl group having 3 to 5 carbon atoms, R^6 is a straight-chain or branched-chain alkyl group having 6 to 11 carbon atoms in the formula (4).

Claim 57 (Previously presented): The process according to claim 56, wherein said alcohol component 1 and said alcohol component 2 have a peroxide value of 1.0 meq/kg or less.

Claim 58 (Previously presented): The process according to claim 57, wherein said alcohol component 1 and said alcohol component 2 have a carbonyl value of 15 or less.

Claim 59 (Previously presented): The process according to claim 56, wherein said esterification reactions in steps (a) and (b) of step (i), are carried out in an inert gas atmosphere or

in an inert gas stream.

Claim 60 (Previously presented): The process according to claim 56, wherein said reaction in step (a) of step (i) is carried out in the absence of a catalyst, and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 61 (previously presented): The process according to claim 56, wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is 10 to 90 mole%, relative to the total amount [(P)+(Q)+(S)+(T)] of alcohol component 1 [(P)+(Q)] used in the first stage esterification reaction and alcohol component 2 [(S)+(T)] used in the second-stage esterification reaction, and

- 1) the entire amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in an amount of 50 mole% relative to the total amount [(P)+(Q)+(S)+(T)] in the first-stage esterification reaction and

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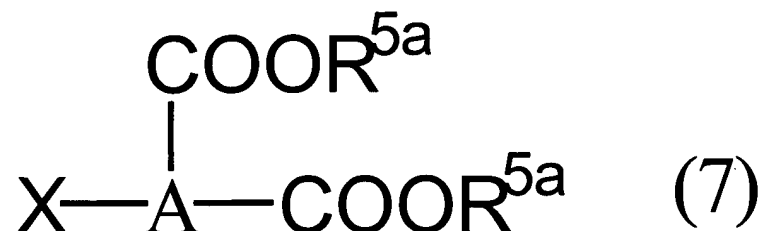
the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S) in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is more than 50 mole% and not more than 90 mole% relative to the total amount [(P)+(Q)+(S)+(T)].

Claim 62 (previously presented): The process according to claim 56, wherein in step (iii), said neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water until the pH of the waste water becomes neutral.

Claim 63 (previously presented): The process according to claim 56, wherein said treatment with adsorbents in step (iv) is carried out using 2 to 4 adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

Claim 64 (Currently amended): A process for preparing an ester mixture, the ester mixture being a mixture of

(1) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7)



wherein A represents a cyclohexane ring[[,]] or a cyclohexene ring or ~~a benzene ring~~, X is a hydrogen atom or methyl, and R^{5a} represents a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and the two -COOR^{5a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A,

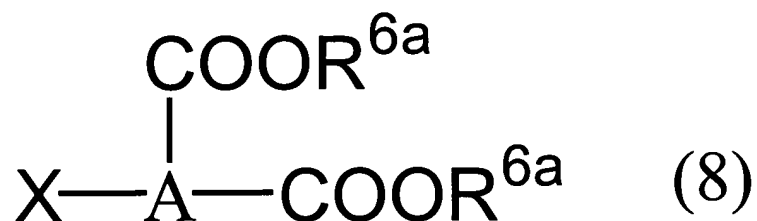
(2) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4)



wherein A and X are as defined in the formula (7), and R⁵ and R⁶ are different from each other and

each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms, and the group --COOR^5 and the group --COOR^6 are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

(3) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl)ester represented by the formula (8)

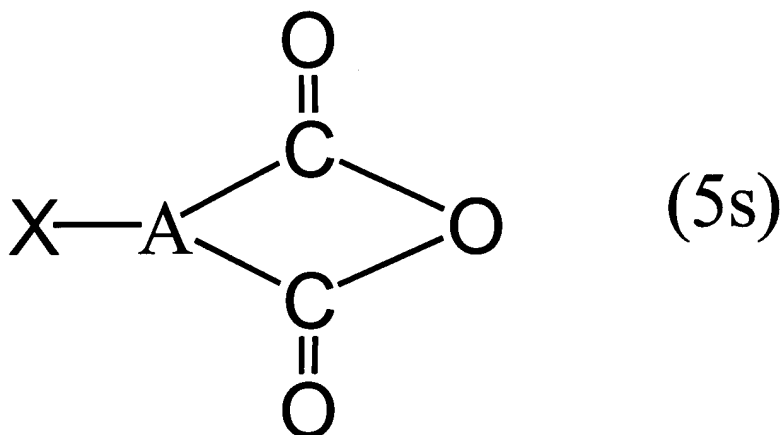


wherein A and X are as defined in the formula (7), and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the two --COOR^{6a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and the ester mixture having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \bullet \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of:

- (i) (a) ~~subjecting an alicyclic or aromatic~~ adjacent dicarboxylic anhydride
represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single

alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A, X and R⁵ are as defined above, and the group -COOR⁵ and the group -COOH are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A,

- (b) subjecting the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst to thereby give a reaction mixture containing said ester mixture of (1) the alicyclic

~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), (2) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4), and (3) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8),

- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
- (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents, and
- (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having the properties 1) to 9).

Claim 65 (Currently amended): The process according to claim 64, wherein the ester mixture contains the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4) under (2) in a proportion of 100, the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7) under (1) in a proportion of 5-300, and the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8) under (3) in a proportion of 7-500, wherein the proportions are expressed in terms of area ratio as determined from a gas chromatogram of the ester mixture.

Claim 66 (Previously presented): The process according to claim 64, wherein the ester mixture is a mixture of an alicyclic adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), an alicyclic adjacent dicarboxylic acid mixed diester represented by the formula (4) and an alicyclic adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8), the ester mixture having a trans isomer/cis isomer ratio of 0/100 to 80/20, wherein the ratios are expressed in terms of area % as determined by gas chromatography.

Claim 67 (previously presented): The process according to claim 64, wherein the monohydric alcohol of 1 to 5 carbon atoms (P) constituting said alcohol component 1 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (Q) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group, and

wherein the monohydric alcohol of 1 to 5 carbon atoms (S) constituting said alcohol component 2 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the

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monohydric alcohol of 6 to 18 carbon atoms (T) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group.

Claim 68 (Previously presented): The process according to claim 64, wherein said alcohol component 1 is a monohydric alcohol of 1 to 5 carbon atoms and said alcohol component 2 is a monohydric alcohol of 6 to 18 carbon atoms.

Claim 69 (Previously presented): The process according to claim 64, wherein said alcohol component 1 and said alcohol component 2 have a peroxide value of 1.0 meq/kg or less.

Claim 70 (Previously presented): The process according to claim 69, wherein said alcohol component 1 and said alcohol component 2 further have a carbonyl value of 15 or less.

Claim 71 (Previously presented): The process according to claim 64, wherein said esterification reactions in steps (a) and (b) of step (i), are carried out in an inert gas atmosphere or in an inert gas stream.

Claim 72 (Previously presented): The process according to claim 64, wherein said reaction in step (a) of step (i) is carried out in the absence of a catalyst, and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C_3 - C_8 alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C_3 - C_{12} fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 73 (Previously presented): The process according to claim 64, wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is 10 to 90 mole%, relative to the total amount [(P)+(Q)+(S)+(T)] of alcohol component 1 [(P)+(Q)] used in the first stage esterification reaction and alcohol component 2 [(S)+(T)] used in the second-stage esterification reaction, and

- 1) the entire amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in an amount of 50 mole% relative to the total amount [(P)+(Q)+(S)+(T)] in the first-stage esterification reaction and the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S)

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in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is more than 50 mole% and not more than 90 mole% relative to the total amount [(P)+(Q)+(S)+(T)].

Claim 74 (previously presented): The process according to claim 64, wherein in step (iii), said neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water until the pH of the waste water becomes neutral.

Claim 75 (previously presented): The process according to claim 64, wherein said treatment with adsorbents in step (iv) is carried out using 2 to 4 adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

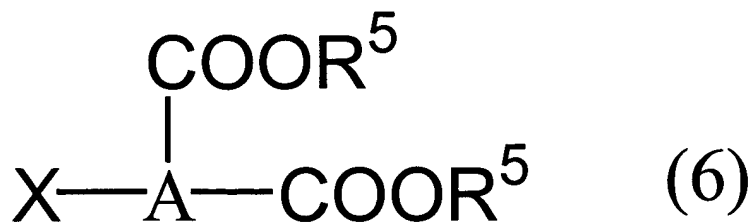
Claim 76 (Currently amended): A process for preparing an ester mixture, the ester mixture being a mixture of:

(1) the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a)



wherein A represents a cyclohexane ring[[,]] or a cyclohexene ring ~~or a benzene ring~~, X is a hydrogen atom or methyl, R^{5a} and R^{6a} are different from each other and R^{5a} represents a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the group -COOR^{5a} and the group -COOR^{6a} are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

(2) an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid diester represented by the formula (6)



wherein A and X are as defined in the formula (4a), and R⁵ is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain

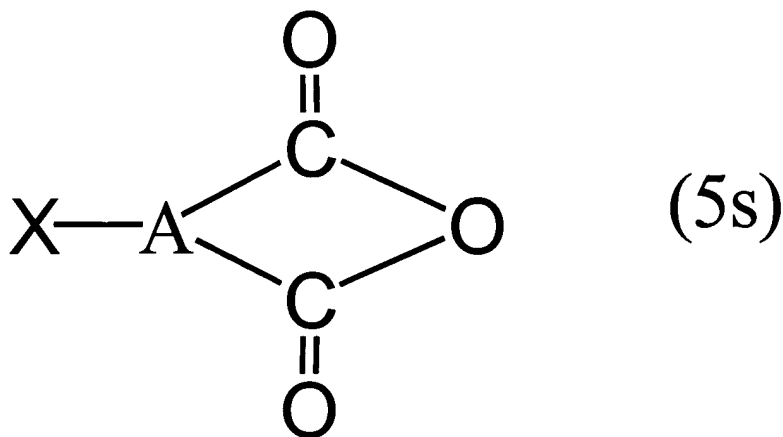
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alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms, and two R⁵ are the same, and the two -COOR⁵ groups are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and the ester mixture having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of:

- (i) (a) subjecting an alicyclic ~~or aromatic~~ adjacent dicarboxylic anhydride represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A, X and R⁵ are as defined above, and the group -COOR⁵ and the group -COOH are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene~~

ring represented by A,

- (b) subjecting the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst to thereby give a reaction mixture containing said ester mixture of the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4) and the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid diester represented by the formula (6),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
- (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents, and
- (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having the properties 1) to 9).

Claim 77 (Previously presented): The process according to claim 76, wherein the monohydric alcohol of 1 to 5 carbon atoms (P) constituting said alcohol component 1 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (Q) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group, and

wherein the monohydric alcohol of 1 to 5 carbon atoms (S) constituting said alcohol component 2 is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (T) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group.

Claim 78 (Previously presented): The process according to claim 76, wherein said alcohol component 1 is a monohydric alcohol of 1 to 5 carbon atoms and said alcohol component 2 is a

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monohydric alcohol of 6 to 18 carbon atoms.

Claim 79 (Previously presented): The process according to claim 76, wherein said alcohol component 1 and said alcohol component 2 have a peroxide value of 1.0 meq/kg or less.

Claim 80 (Previously presented): The process according to claim 79, wherein said alcohol component 1 and said alcohol component 2 further have a carbonyl value of 15 or less.

Claim 81 (Previously presented): The process according to claim 76, wherein said esterification reactions in steps (a) and (b) of step (i), are carried out in an inert gas atmosphere or in an inert gas stream.

Claim 82 (Previously presented): The process according to claim 76, wherein said reaction in step (a) of step (i) is carried out in the absence of a catalyst, and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

Claim 83 (Previously presented): The process according to claim 76, wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is 10 to 90 mole%, relative to the total amount [(P)+(Q)+(S)+(T)] of alcohol component 1 [(P)+(Q)] used in the first stage esterification reaction and alcohol component 2 [(S)+(T)] used in the second-stage esterification reaction, and

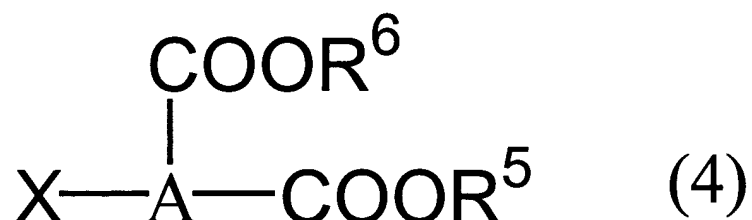
- 1) the entire amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in an amount of 50 mole% relative to the total amount [(P)+(Q)+(S)+(T)] in the first-stage esterification reaction and the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S) in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is more than 50 mole% and not more than 90 mole% relative to the total amount [(P)+(Q)+(S)+(T)].

Claim 84 (Previously presented): The process according to claim 76, wherein in step (iii), said neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water

until the pH of the waste water becomes neutral.

Claim 85 (Previously presented): The process according to claim 76, wherein said treatment with adsorbents in step (iv) is carried out using 2 to 4 adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, non-sulfonic acid ion exchange resins and synthetic hydrotalcite.

Claim 86 (Currently amended): A process for preparing an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4)



wherein A represents a cyclohexane ring[[,]] or a cyclohexene ring ~~or a benzene ring~~, X is a hydrogen atom or methyl, R⁵ and R⁶ are different from each other and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon

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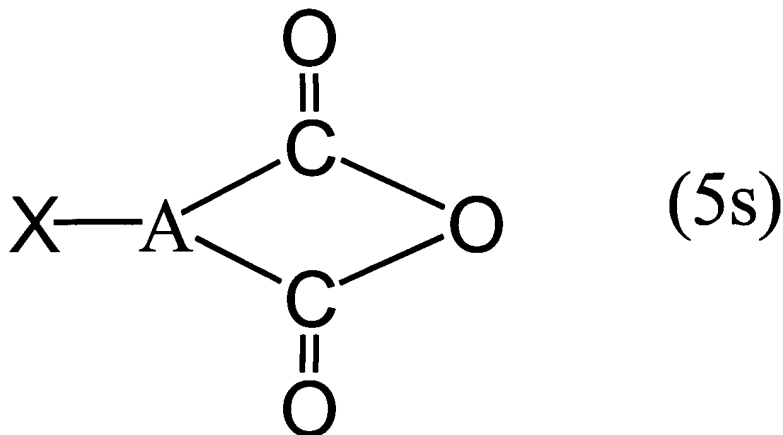
atoms, and the group --COOR^5 and the group --COOR^6 are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester having the following properties:

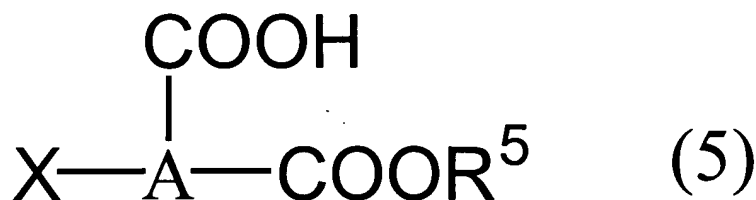
- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of:

- (i) (a) subjecting an alicyclic ~~or aromatic~~ adjacent dicarboxylic anhydride represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



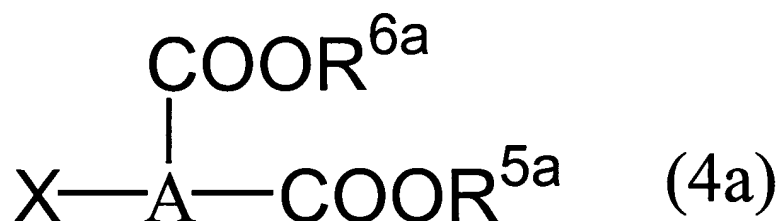
wherein A, X and R⁵ are as defined above, and the group -COOR⁵ and the group -COOH are

attached to two adjacent carbon atoms of the cyclohexane ring[[,]] ~~or cyclohexene ring or benzene ring~~ represented by A,

- (b) subjecting the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst to thereby give a reaction mixture containing an ester mixture including the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
- (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents,
- (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having the properties 1) to 9), and
- (vi) separating the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4) from the ester mixture to thereby give the alicyclic ~~or aromatic~~ adjacent

dicarboxylic acid mixed diester represented by the formula (4).

Claim 87 (Currently amended): A process for preparing an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a)



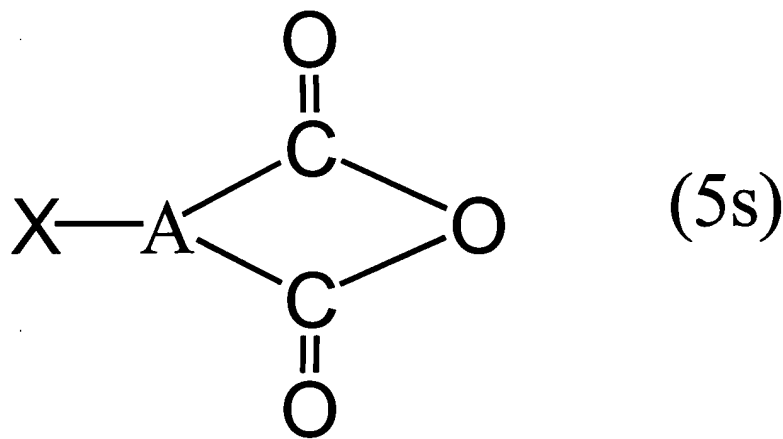
wherein A represents a cyclohexane ring[[,]] or a cyclohexene ring ~~or a benzene ring~~, X is a hydrogen atom or methyl, R^{5a} and R^{6a} are different from each other and R^{5a} is a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the group –COOR^{5a} and the group –COOR^{6a} are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A, and

the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester having the following properties:

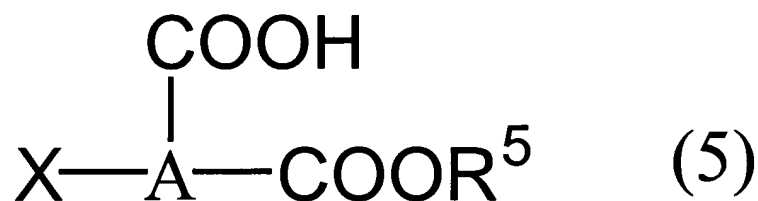
- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of:

- (i) (a) subjecting an alicyclic ~~or aromatic~~ adjacent dicarboxylic anhydride represented by the formula (5s)



wherein A and X are as defined above and alcohol component 1 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio), to esterification reaction to thereby give an alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A and X are as defined above, and R⁵ is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms, and the group -COOR⁵ and the group -COOH are attached to two adjacent carbon atoms of the cyclohexane ring[[,]] or cyclohexene ring ~~or benzene ring~~ represented by A,

- (b) subjecting the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and alcohol component 2 which is a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio), to a further esterification reaction in the absence

- of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst
- to thereby give a reaction mixture containing an ester mixture including the alicyclic
- ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the ester mixture in a crude form,
 - (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
 - (iv) purifying the crude ester mixture neutralized and washed with water in step (iii) by treatment with 1 to 4 adsorbents,
 - (v) dehydrating the ester mixture purified in step (iv) to thereby give the ester mixture having the properties 1) to 9), and
 - (vi) separating the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a) from the ester mixture to thereby give the alicyclic ~~or aromatic~~ adjacent dicarboxylic acid mixed diester represented by the formula (4a).

Claim 88 (Previously presented): The process according to claim 86, wherein R^5 is a straight-chain alkyl group having 1 to 5 carbon atoms or a branched-chain alkyl group having 3 to 5 carbon atoms, R^6 is a straight-chain or branched-chain alkyl group having 6 to 11 carbon atoms in the formula (4).